

The dependence of the critical energy density and hot-spot temperature on the radius of metal nanoparticles in PETN

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The dependencies of critical energy density and corresponding hot-spot temperature were calculated in terms of thermal model of energetic materials laser initiation for 12 metal nanoparticles in pentaerythritol tetranitrate (PETN) at pulse duration 12 ns. We showed that the critical hot-spot temperature depends mostly on the nanoparticle's radius while its dependence on the specific heat of the metal is much weaker. The equations for the critical parameters of initiation on radius and specific heat of the nanoparticles were derived. The results are essential for the explosive compounds for optical detonator cup optimization.

Keywords: pentaerythritol tetranitrate, metal nanoparticles, laser radiation, hot-spot model.

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1. Introduction

The use of pulsed laser methods for the initiation of the explosive processes is under consideration now as a promising replacement for electrical methods of initiation. The main reason is the crucial increasing of safety in explosives utilization and the reduction and minimization of environmental risks and technological disasters. The use of explosives drastically lowers the costs for tunneling works, which determines the increasing scale of their utilization [1]. The most intensively developed way to improve the safety of storage, transportation and use of explosives is the development of energetic materials selectively sensitive to the needed impact [1,2]. Optical detonators based on silver azide [3,4] have minimum thresholds value when initiated with laser pulse, but they are also sensitive to other types of impact (heat, electric spark, etc.). To create selectively sensitive materials to laser radiation, one introduces light-absorbing nanoparticles [5] or the synthesis of new compound with absorption bands matching the wavelength of the radiation sources to the existing transparent explosives [6–8]. The minimum initiation energy density of explosive decomposition of the regular blasting explosives (PETN and RDX) with additives of aluminum [2,9,10], nickel [10,11] and other metals nanoparticles is of the order of 1 J/cm², which is more than hundred times less than for pure pressed tablets of this explosives. This is an energy saving method to initiate due to the concentration of thermal energy in "hot-spots" and the implementation of thermal explosion in a microcenter variant. The initiation mechanism based on the energy absorption by small inclusions in the explosives volume was formulated in [12] for heavy metal azides. In [13–15], the model was modernized with optical properties and melting processes of nanoparticles and the matrix. The relevance of modernization and study of the hot-spot model is defined by its fundamental value for the development of the mechanisms of thermal explosion model representations, as well as by the applied one based on the selectively sensitive optical detonator creation.

The purpose of this work is the determination of the critical parameters for the initiation of explosive decomposition reaction calculation in the framework of the hot-spot model of thermal explosion, defining the role of the thermophysical parameters of the nanoparticles' material at the stage of formation and development of the reaction hot-spot and analysis of the results in dimensionless coordinates for the process. The compounds based on regular secondary explosives PETN and the nanoparticles of 12 metals were selected as model systems.

The process of initiating self-accelerating decomposition in transparent explosives and metal nanoparticles of different natures can be divided into several stages: the transfer of energy from the output window of the source of monochromatic radiation to nanoparticles, the absorption of the pulse energy of nanoparticles, heating the nanoparticles and labile matrix with the formation of the explosive decomposition hot-spot. The influence of phase transitions in the nanoparticle and the matrix were considered in [15], where it was shown that the melting has little effect on threshold values for initiation of PETN – aluminum nanoparticle systems. Depending on the material and radius of nanoparticles, the absorption efficiency varies considerably [14,15]. The consideration of

multiple scattering in the sample leads to a substantial increase of illumination in the sample's volume [17–19]. The values of the reflection coefficient from the front surface of the sample, experimentally defined in [20], also change significantly depending on sample parameters and radiation wavelength. The optical effect consideration greatly complicates the mechanism for explosive decomposition, but it is possible to allocate a relatively slow subsystem: heating of the nanoparticles where the ratio of the absorption efficiency coefficients of the nanoparticles is 1 [12], conductive heat transfer and thermal chemical decomposition of explosives. It is rational to take into account the peculiar features of nearly instantaneous propagation and absorption of laser light by metal nanoparticles by later correcting the calculated dependence of the nanoparticles' radii critical parameters on previously defined coefficients (as in [15, 21, 22]).

Additional assumptions implicitly used by the authors of the hot-spot model [12, 17] are: metal nanoparticles do not have an oxide film; they are in tight contact with the matrix explosives; and do not interact with it chemically. The spherical symmetry originally used by the authors of the model, is due to three circumstances. First, in the case of pressed pellets of explosives, the light falling onto the sample undergoes multiple reflection on the grain boundaries and metal nanoparticles [17–19]. Because of the randomness of the reflection acts and the significantly negative values of the mean cosine of the scattering angle by metal nanoparticles in PETN [23], the light intensity averages over every direction. Second, the absorbed pulse energy, turning into heat, quickly spreads over the nanoparticles' volume, especially when they are not very big (less than 120 nm). The values of the thermal diffusivity of metals are much higher than that of PETN, which lead to almost uniform heating of the nanoparticles during the pulse duration (~ 10 ns) [24]. Third, with one-dimensional approach it is possible to produce the necessary mathematical calculations with much greater accuracy for a reasonable time. Thus, the use of spherical symmetry is justified. The system of differential equations describing the processes of conductive heat transfer, and heat generation due to chemical decomposition of energetic material, has the form [12]:

$$\begin{aligned} \frac{\partial T}{\partial t} &= \alpha \left(\frac{\partial^2 T}{\partial x^2} + \frac{2}{x} \frac{\partial T}{\partial x} \right) + \frac{Q}{C} k_0 n \cdot \exp \left(-\frac{E}{k_B T} \right), & x > R, \\ \frac{\partial n}{\partial t} &= -k_0 n \cdot \exp \left(-\frac{E}{k_B T} \right), & x > R, \\ \frac{\partial T}{\partial t} &= \alpha_m \left(\frac{\partial^2 T}{\partial x^2} + \frac{2}{x} \frac{\partial T}{\partial x} \right), & x < R, \end{aligned} \quad (1)$$

where T is temperature, n is relative concentration of explosives decreases in the course of the decomposition reaction from 1 to 0, $\alpha = 1.1 \cdot 10^{-3} \text{ cm}^2 \text{ s}^{-1}$ and α_m are thermal diffusivities of the matrix and nanoparticle, k_B is Boltzmann constant, $E = 165 \text{ kJ}/(\text{mol} \cdot \text{K})$ is energy of activation, $Q = 9.64 \text{ kJ}/\text{cm}^3$ is heat efficiency of the decomposition, $k_0 = 1.2 \cdot 10^{16} \text{ s}^{-1}$ is pre-exponential factor [14], $C = 2.22 \text{ J}/(\text{cm}^3 \text{ K})$ is volumetric heat capacity of the explosives with boundary condition for $x = R$ is:

$$J - C_m \alpha_m \cdot \left. \frac{\partial T}{\partial x} \right|_{x \rightarrow R-0} + C \alpha \cdot \left. \frac{\partial T}{\partial x} \right|_{x \rightarrow R+0} = 0, \quad (2)$$

where C_m is volumetric heat capacity of the nanoparticle, $J(t)$ is the absorbed density of the laser pulse radiation power. In terms of the model we neglecting the possible non-ideal thermal contact of the nanoparticle and the explosive [25] and possible diffusion of the explosive material [26].

The facilities based on a neodymium laser typically used for initiation of explosive decomposition have the dependence of the radiation power of the pulse close to the normal distribution function [27]. Taking the maximum intensity of the pulse as the reference time position, we obtain for the quantity $J(t)$ the expression:

$$J(t) = k_i H_0 \cdot \exp(-k_i^2 t^2) / \sqrt{\pi}, \quad (3)$$

where $k_i = \frac{2\sqrt{\ln(2)}}{t_i}$ is a parameter that determines the pulse duration; H_0 – the pulse energy density. Multipliers of equation (3) normalized the integral of $J(t)$ with respect to time on H_0 . Model of explosive decomposition under initiation by a laser pulse is rational initially to explore in dimensional variables, with a further choice of dimensionless. To determine the critical parameters of explosive decomposition, the system of equations for the model (1-3) was numerically solved on the grid with a variable coordinate step by the method described in [24, 28].

The calculations were performed for 12 metals to elucidate the role of thermophysical properties for the material nanoparticles. The values of heat capacities and thermal conductivities of the metals used were taken from [21]. The variation of the nanoparticles radius was carried out over the 10–120 nm range. This interval covers the radii used in the experimental studies of aluminum [2, 19] and nickel [10, 11] nanoparticles, a typical area radii of noble

metal nanoparticles, 10–20 nm, by effectively absorbing electromagnetic radiation at the frequencies of plasmon resonance [22, 29] and relatively large radii of transition metals at which the radiation of the first harmonic of a neodymium laser is absorbed the most efficiently [23] though the plasmon band is observed in the UV region for aluminum nanoparticles [30].

2. The method of calculation and results

The calculation of the critical energy density was carried out using bisection method with the interval reduction achieving up to a relative accuracy of 10^{-8} . The determination of the system bifurcation point (1)–(3) with such precision requires the minimization of errors associated with the calculation of critical parameters for explosive decomposition initiation and definition of the explosive decomposition parameters close to the bifurcation point [31]. For further analysis, the maximum energy density at which the explosion was not observed H was retained. To consider the contribution of the chemical reaction in the threshold of initiation, the maximum temperature on the surface of the nanoparticles with the energy density $H(T_{max})$ and the time of its occurrence $t(T_{max})$ were calculated. The calculation of the maximum temperature T_{max0} for each system at the energy density of H without taking into account the heat flux due to chemical decomposition (installed $k_0 = 0$) and the time of appearance of maximum temperature $t(T_{max})$ was subsequently performed.

For 12 selected metals in PETN, we calculated selected characteristics of the process near the bifurcation point. Table 1 presents the results of calculations of H , T_{max} , $t(T_{max})$, T_{max0} and $t(T_{max0})$ with the radii of nanoparticles of cobalt ranging from 10–120 nm. The critical energy density values significantly depend upon the nanoparticles' radii, but an even more amazing result is the significant dependence of the temperature characteristic of the process on the radius of the nanoparticles. From Table 1, it follows that T_{max} is significantly higher than T_{max0} . The temperature difference increases with decreasing nanoparticle radius. For cobalt nanoparticles with a 10 nm radius and a pulse duration of 12 ns, the difference is around 346 K and in the limit of large radius is less than 40 K. The dependence $T_{max0}(R)$ is well described by the equation:

$$T_c(R) = T_\infty + \frac{r_T}{R}, \quad (4)$$

where the variable parameters T_∞ and r_T (4) are determined with approximation of the corresponding dependencies. The values of the variable parameters for the 12 considered metals in the matrix PETN are given in Table 2.

As one can see, the values of T_∞ for the 12 metals differ by only 19 K. The parameter r_T deviates maximally from the middle value by 2% only. Consequently, the critical temperature of the reaction hot-spot of explosive decomposition is almost independent on the nature of the metal nanoparticles.

Let us consider the dependence of the critical energy density of the radius and the nature of the nanoparticles. Consider a simplified model for thermophysical processes with heating of nanoparticles in inert matrices. The maximum change by primary hot-spot temperature without considering contribution of chemical decomposition of the matrix could be estimated with the expression [11, 12]:

$$\Delta T = \frac{3}{4\pi} \cdot \frac{\pi R^2 H}{C_m R^3 + C((R+h)^3 - R^3)}. \quad (5)$$

In the numerator of formula (1), the energy absorbed by the nanoparticle as a result of the pulse action remains, and the denominator estimates the overall heat capacity of the system, involving heating of the matrix layer of thickness h and nanoparticles to the same temperature. The value h is the effective thickness of the heated layer of the matrix, determined by the pulse duration, heat capacity and thermal diffusivity of the matrix, but not the thermophysical properties of the nanoparticles (as the ration $\alpha \ll \alpha_m$ is fulfilled for all the metals). Using the formula (4), approximating the dependence of the critical temperature of the hot-spot on the nanoparticles' radii without a chemical reaction as T_{max0} , and dropping off the term proportional to h^3 , we get:

$$H = \frac{4}{3R} \cdot \left(T_\infty + \frac{r_T}{R} - T_0 \right) \cdot [C_m R^2 + 3Ch \cdot (R+h)]. \quad (6)$$

Approximation of the calculated dependences $H(R)$ by formula (6) was performed for all 12 selected metals in PETN applying the previously defined fitting parameters T_∞ and r_T that are given in columns 2 and 3 of Table 2. The array h is shown in column 4 of Table 2. From the estimated values of h , it follows that they are independent of the nature of the metal nanoparticles: the average value of $h = 34.87$ nm and a maximum deviation is only 0.5 %. This result is consistent with its physical meaning as the typical thickness of the heated layer.

Using formula (6) we could demonstrate the effect of the metals' specific heat on the dependence of $H(R)$. The coordinates of the minima for the dependences $H(R)$ for 12 metals in PETN (H_{min} and R_{min}) are presented

TABLE 1. Calculated with the radii of the cobalt nanoparticles R values H , T_{max} , $t(T_{max})$, T_{max0} , $t(T_{max0})$

R , nm	H , mJ/cm ²	T_{max} , K	$t(T_{max})$, ns	T_{max0} , K	$t(T_{max0})$, ns
10	136.170749	1497.06	4.0496	1150.90	1.3178
15	97.928431	1360.16	6.080	1107.83	1.9417
20	81.485410	1289.6	7.5321	1088.68	2.5232
25	72.908448	1245.46	8.6159	1077.56	3.0538
30	68.110572	1214.96	9.4775	1070.86	3.4867
35	65.3497462	1192.23	10.200	1066.07	3.8931
40	63.8247203	1174.39	10.722	1062.41	4.2578
45	63.1029816	1159.97	11.203	1059.51	4.6208
50	62.931098	1148.01	11.495	1057.11	4.9111
55	63.1510649	1137.91	11.601	1055.09	5.2318
60	63.6594571	1129.31	11.725	1053.32	5.4625
65	64.3858326	1121.89	11.692	1051.77	5.7452
70	65.2806344	1115.45	11.606	1050.37	5.9199
75	66.3080204	1109.81	11.596	1049.10	6.138
80	67.4414514	1104.82	11.558	1047.95	6.3609
85	68.6608745	1100.38	11.488	1046.87	6.5691
90	69.9508670	1096.38	11.465	1045.86	6.6802
95	71.2993787	1092.77	11.388	1044.92	6.8438
100	72.6968651	1089.47	11.400	1044.04	7.0616
105	74.1356704	1086.45	11.316	1043.19	7.2034
110	75.6095868	1083.66	11.349	1042.39	7.3449
115	77.1135275	1081.08	11.330	1041.62	7.4714
120	78.6438735	1078.68	11.316	1040.86	7.6307

in the last columns of Table 2. Assuming that the dependence $T_{max0}(R)$ at relatively large radii R_{min} corresponding to the minimum H , is weak, in this limit, the dependence $H(R)$ will match the radius of most heated nanoparticles [14]:

$$R_{min} = \sqrt{\frac{6\alpha k_i C}{C_m}}. \quad (7)$$

Equation (7) leads to a linear dependence of the R_{min} square on the reverse value of the metal heat capacity (C_m). Fig. 1 shows the dependence of the abscissa of the minimum R_{min} as a dependence on the metal volume specific heat capacity in the rectifying coordinates. Points in the figure show the data for the metals lead, tin, aluminum, silver, gold, palladium, vanadium, chromium, copper, iron, cobalt, nickel, and their good linear approximation.

TABLE 2. Calculated critical parameters of explosive decomposition for PETN with various metal nanoparticles

Metal	T_{∞} , K	r_T , K·nm	h , nm	H_{min} , mJ/cm ²	R_{min} , nm
Pb	1070.45	1638.38	35.03	47.712	79.20
Sn	1071.39	1644.82	34.98	49.430	74
Al	1074.31	1688.38	34.85	54.987	61.3
Ag	1074.35	1689.10	34.85	55.206	60.90
Au	1074.43	1690.08	34.85	55.352	60.60
Pd	1075.68	1703.11	34.81	58.068	56
V	1076.33	1709.01	34.79	58.559	55.20
Cr	1076.03	1704.22	34.81	59.930	53.30
Cu	1075.99	1700.82	34.83	61.145	51.60
Fe	1076.73	1702.26	34.82	61.802	50.80
Co	1076.41	1698.39	34.84	62.928	49.40
Ni	1076.51	1693.51	34.85	64.097	48.10

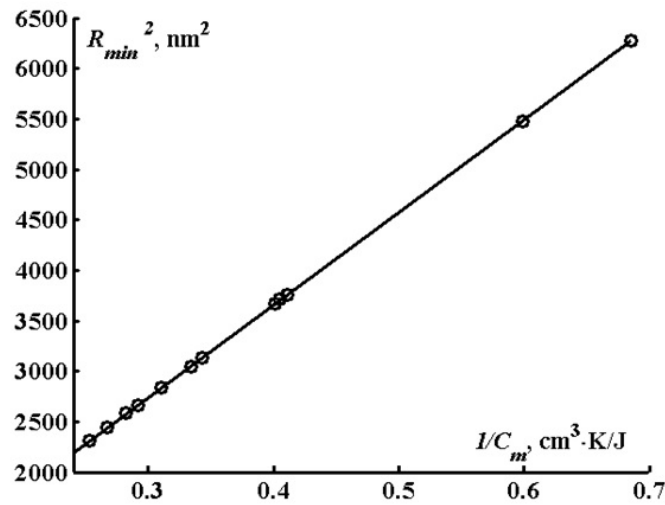


FIG. 1. The dependence of the square R_{min} , from the reverse value of the metal heat capacity (C_m) with pulse width was 12 ns. The dots are the data for the metals: lead, tin, aluminum, silver, gold, palladium, vanadium, chromium, copper, iron, cobalt, nickel. Line-approximation of the line passing through the origin

3. Conclusion

Using the characteristics of 12 metals in the present work allowed us to investigate the effect of nanoparticles' specific heat in the range from 1.46 (lead) to 3.96 J/(cm³·K) (nickel) on the main characteristics of the process of laser initiation. The obtained analytical equations quantitatively describing the results of the calculations allow us to predict the influence of specific heat over a wide range that it is necessary to expand the range of materials considered as promising additives for the selective sensitivity of explosives by laser radiation.

Currently, the refinement of the hot-spot model for the laser initiation of thermal explosion of energetic materials containing metal nanoparticles occurs in a number of directions that span the optical and thermophysical processes. For this reason, the comparison with experiment of the results of this study is difficult. Previously it was shown [9] that taking into account the optical properties of individual metal nanoparticles allows us to

reduce considerably the differences of the absolute theoretical and experimental values for the critical energy density. The last quantity can be reliably predicted if we know the kinetic parameters for the decomposition reaction in the temperature region in which a hot-spot is formed. Direct measurement of the rate constant over this temperature range is difficult, therefore, reasonable extrapolations from the low temperature region generally used may not always be applicable. Alternatively, the obtained expressions allow one to predict correctly the effect of the thermophysical parameters of nanoscale additives, which is essential in the development of new energetic materials.

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